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Oscillator strength measurements of the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ Rydberg transitions of strontium

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We report the experimentally determined oscillator strengths for the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ Rydberg transitions of strontium using two-step excitation in conjunction with a thermionic diode ion detector. The absolute photoionization cross section from the $5s6s\ ^1S_0$ excited state has been determined by adjusting the polarization vector of the ionizing laser beam parallel, perpendicular, and at the magic angle with respect to that of the exciting dye laser. The measured absolute value of the photoionization cross section 0.9 ± 0.2 Mb at the 5s threshold is used to extract the f values of the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ ($26 \leq n \leq 73$) Rydberg transitions. The oscillator strength in the discrete region merges smoothly to the oscillator strength density at the ionization threshold.

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I. INTRODUCTION

The valence electrons of neutral strontium can be excited to higher Rydberg states to explore spectroscopic parameters such as photoionization cross section and oscillator strength. The oscillator strength of a transition is normally measured by comparing different transitions of known number density and absolute intensity. It can also be measured using the lifetime of an excited state where a single decay transition channel exists. Over the past several decades, theoretical as well as experimental determinations of optical oscillator strengths of Rydberg transitions, particularly from the ground state, have been reported but little data are available from the excited states. Wiese [1] and Huber and Sandeman [2] reviewed the experimental and theoretical techniques for the measurement of oscillator strengths of different transitions. On the theoretical side, Hansen [3] presented the f values for the $5s^2\ ^1S \rightarrow 5s5p\ ^1P$ and $5s4d\ ^1D \rightarrow 5s5p\ ^1P$ transitions of strontium both in the dipole length and velocity approximations. Hunter and Berry [4] calculated the oscillator strengths of the atomic transitions of alkaline earth atoms using configuration-interaction and electronic rotor-vibrator wave functions and compared the theoretical and experimental results. Barrientos and Martin [5] calculated the oscillator strengths and photoionization cross sections of the lower members of the principal series of strontium. Vaeck *et al.* [6] calculated the oscillator strength and level energies for the singlet transitions and Werij *et al.* [7] inferred the disagreement between the experimental and theoretical oscillator strengths in strontium. They used R -matrix, multi-channel quantum defect theory (MQDT), and modified Coulomb-approximation calculations along with branching ratio mea-

surements to get transition rates for the allowed singlet and triplet transitions. Migdalek and Stanek [8] used relativistic multiconfiguration Dirac-Fock method and determined transition energies and oscillator strengths for the $5s^2\ ^1S_0 \rightarrow 5s5p\ ^{1,3}P_1$ transitions of strontium and its isoelectronic sequence.

Experimentally, Penkin and Shabanova [9] measured relative as well as absolute oscillator strengths of the principal series of strontium and barium via Hook method and also reported oscillator strengths for the $^3P_{0,1,2} \rightarrow ^3S_1$ transitions of strontium and $^3P_{0,1,2} \rightarrow ^3D_{1,2,3}$ transitions of magnesium. Parkinson *et al.* [10] using the Hook method determined oscillator strength distribution in the discrete and continuum spectra of the principal series of calcium, strontium, barium and extracted photoionization cross section at their first ionization thresholds. Lu [11] studied the same series in strontium and derived the f values distribution in the discrete and in the autoionization region using two-channel quantum defect theory. Ueda *et al.* [12] used the Hook method to measure relative as well as absolute oscillator strengths from the $5s5p\ ^3P_{0,1,2}$ levels of strontium and other alkaline earths. Garton *et al.* [13] measured relative f values of the $5snp\ ^1P_1$ states with the help of magneto-optical rotation exploiting the polarization property of the synchrotron radiation. Subsequently, Connerade *et al.* [14] measured absolute oscillator strengths of the principal series ($20 \leq n \leq 43$) of strontium and barium ($15 \leq n \leq 42$) using Faraday rotation technique in conjunction with a pulsed magnet and a dye laser system. Mende and Kock [15] determined the f values of the $5s5p\ ^1P_1 \rightarrow 5snd\ ^1D_2$ ($30 \leq n \leq 60$) transitions of strontium and $5s6p\ ^1P_1 \rightarrow 6snd\ ^1D_2$ ($31 \leq n \leq 60$) transitions of barium using absolute value of the photoionization cross section measured at the series limit. Subsequently, Mende and Kock [16] determined oscillator strength of the principal series of strontium using the measured photoionization cross section at threshold (12 ± 2.5) Mb to calibrate the f values up to $n = 60$ on an absolute scale. The results were compared with that of Connerade *et al.* [14] and possible reasons of deviations from their results were specified. Brownutt *et al.* [17] demonstrated the controlled photoionization loading of $^{88}\text{Sr}^+$ for precision ion trap experiments and Matsuo *et al.* [18] reported the photoionization characteristics of strontium in to

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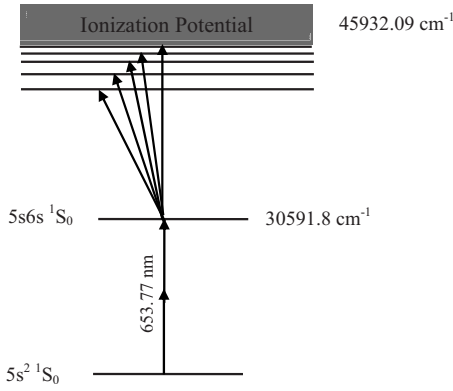


FIG. 1. A schematic energy level diagram of strontium showing relevant levels in the excitation process. The level energies are taken from the NBS Tables (NBS, 2008, Moore, 1971) [41].

$5skl$ continua through the spin resolved ion detection by laser induced fluorescence. The photoionization cross section from the $5s5p\ ^1P_1$ and $5s5p\ ^3P_1$ excited states of strontium at the first ionization threshold was reported by Haq *et al.* [19] as 11.4 ± 1.8 Mb and 10.7 ± 1.7 Mb, respectively, which was subsequently used to determine the oscillator strengths of the $5snd\ ^1D_2$ ($19 \leq n \leq 68$) and the $5snd\ ^3D_2$ ($24 \leq n \leq 70$) series. Recently, Lontos *et al.* [20] reported the single and double ionization of strontium in the vicinity of four photon excitation of the $5p^2\ ^1S_0$ doubly excited state.

The objective of the present study is to extend the knowledge of photoionization cross sections to other excited states of strontium and to determine oscillator strengths of the transitions originating from these excited states. In this work, the photoionization cross section (as a prerequisite for oscillator strength measurement) has been determined at the $5s$ ionization threshold by ionizing laser at 651.8 nm using linearly polarized light with the polarization vectors parallel, perpendicular and at the magic angle with respect to the exciting laser beam. The measured value of photoionization cross section is then used to determine f values of the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ ($26 \leq n \leq 73$) Rydberg transitions.

II. EXPERIMENTAL DETAILS

The experimental arrangement to measure oscillator strengths of the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ Rydberg series of strontium is the same as described in our earlier papers [19,21–23]. In brief, strontium vapors were confined in a thermionic diode ion detector. Argon at a pressure of ~ 1 Torr was used as a buffer gas. About 20 cm central portion of the thermionic diode was heated by a clamp-shell furnace, operated at ≈ 920 K corresponding to ≈ 0.2 Torr strontium vapor pressure [24]. Adjusting the cathode current and the buffer gas pressure in the cell, the ionization signal was optimized such that the detector works in the linear mode, i.e., the ionization signal increases linearly with the increase of the ionizing laser energy density.

Following the excitation schemes shown in Fig. 1, a Hanna-type dye laser [25], charged with DCM-640 dye and pumped by the SHG of a Nd:YAG laser (Quantel, France)

was tuned at 653.77 nm to populate the $5s6s\ ^1S_0$ intermediate state via two-photon excitation from the ground state. The excited atoms were then ionized by the PDL-2 dye laser pumped by the SHG of the same Nd:YAG laser, charged with DCM-640 and tuned at 651.8 nm, which corresponds to the $5s$ ionization threshold wavelength. At the ionizing laser wavelength, the ionization signal was registered as a change in the voltage across a 100 k Ω load resistor. The intensity of the ionizing laser was varied by inserting neutral density filters (Edmund Optics, USA) and on each insertion the energy was measured by an energy meter (R-752, Universal Radiometer). The variation in the amplitude of the ionization signal with laser intensity was recorded using a 200 MHz digital storage oscilloscope (TDS 2024) and a computer through RS 232 interface.

The $5snp\ ^1P_1$ states were excited by scanning the second dye laser in the wavelength range from 651 to 662 nm. The wavelength calibration was achieved by recording simultaneously the output from the thermionic diode, the optogalvanic spectra from the neon hollow cathode lamp and rings from a 1-mm-thick fused silica Fabry-Perot etalon (FSR 3.33 cm^{-1}) via three boxcar averagers (SR 250). The optogalvanic signals from the hollow cathode lamp provided well-distributed spectral lines of neon in the region of interest [26] that served as wavelength markers. The interference fringes from the etalon were used to interpolate between the neon lines. The spectrum was recorded with a 0.02 cm^{-1} scanning step of the dye laser. From the location of the peak signal positions, the transition energies have been determined within an accuracy of about $\pm 0.2\ \text{cm}^{-1}$. The linewidth of the dye laser was $\leq 0.3\ \text{cm}^{-1}$ and both the laser pulses were linearly polarized. The exciting and the ionizing laser beams entered the thermionic diode from the opposite sides and overlapped spatially and temporally at the center. The relative delay between the exciting and the ionizing laser pulses was controlled by an optical delay line.

III. RESULTS AND DISCUSSION

The excitation scheme used to determine the optical oscillator strength of the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ Rydberg series is shown in Fig. 1. The technique adopted for the measurement of oscillator strength in the present work is similar to that developed by Mende and Kock [15] which has been used to study oscillator strengths of Rydberg transitions of various atoms [21–23]. In this technique the measured photoionization cross-section value at the threshold is used to determine oscillator strength of the transition at the corresponding wavelength using the relation

$$f_n = \frac{mc}{\pi} \left(\frac{4\pi\epsilon_0}{e^2} \right) \left(\frac{S_n}{S_{1+}} \right) \left(\frac{\lambda_{1+}}{\lambda_n} \right) \sigma^{1+}. \quad (1)$$

Here f_n is the oscillator strength for the n th transition of a Rydberg series, which is directly proportional to the photoionization cross section σ^{1+} , measured at a wavelength λ_{1+} , i.e., at the ionization threshold. The S_{1+} is the ion signal at the ionization threshold and S_n is the integrated ion signal intensity for the n th transition. The parameter σ^{1+} is the absolute photoionization cross section from the $5s6s\ ^1S_0$ ex-

cited state at the $5s$ ionization threshold, which has been determined using the saturated ionization technique [27–32]. In this experimental technique, the photoions produced as a result of two-step photoionization process are detected as a voltage drop across the load resistor in the ion detector. The collected charge Q per pulse in terms of the voltage signal is represented as

$$Q = \left(\frac{\text{Voltage signal}}{R} \right) \Delta t = e N_0 V_{\text{vol}} \left[1 - \exp \left(- \frac{\sigma U}{2 \hbar \omega A} \right) \right]. \quad (2)$$

Here R is the load resistance and Δt (in seconds) is the full width at half maximum of the photoion signal, e (in C) is the electronic charge, N_0 (cm^{-3}) is the density of the excited atoms, A (cm^2) is the cross-sectional area of the ionizing laser beam, U (in J) is the total energy per ionizing laser pulse, $\hbar \omega$ (in J) is the energy per photon of the ionizing laser beam, V_{vol} (cm^3) is the laser interaction volume and σ (cm^2) is the absolute cross section for photoionization [28,33]. The photoionization cross section depends upon the curvature of the curve of the ionization signal versus ionizing laser energy, whereas the density of the excited state is independent of the cross section. The spatial intensity distributions of the exciting and the ionizing laser beams were observed by scanning a p - i - n photodiode across both the axes and were found to be Gaussian like distribution. The spot sizes were determined at which the irradiance (intensity) falls to $1/e^2$ of its axial value. The diameter of the exciting laser was ≈ 4 mm, and the ionizing laser was confined to ≈ 2 mm by passing through an iris. Since the diameter of the ionizing laser beam was smaller than that of the exciting laser, the spatial overlap of the beams was easy to achieve. The ionizing laser beam was focused by a long focal length lens to meet the power density requirements for saturation and to further ensure the spatial overlap of both the laser beams. The area of the overlapping region in the confocal limit was calculated using the method described by Demtröder [34] and Song *et al.* [35]. Incorporating all these requirements and under the best alignment conditions, we have measured the absolute photoionization cross section from the $5s6s \ ^1S_0$ excited state at the $5s$ ionization threshold.

A typical experimental data points for the photoionization from the $5s6s \ ^1S_0$ excited state, ionizing laser energy versus the photoionization signal, and the fitted curve of Eq. (2) is presented in Fig. 2. The curvature of the fitted curve is small and there is no clear indication of saturation even at the higher ionizing laser intensity indicating very small value of the cross section from this state. The best fit to the experimental data yields the photoionization cross section as $\sigma = 0.9$ (0.2) Mb. In this experiment, the excitation laser corresponding to the $5s^2 \ ^1S_0 \rightarrow 5s6s \ ^1S_0$ was linearly polarized with the polarization vector along the direction of propagation. The total angular momentum of the excited state is $J = 0$ therefore the $M_J = 0$ sublevel is populated. The polarization vector of the ionizing laser was set parallel to the exciting laser which accessed the $M_J = 0$ out of the $M_J = 0, \pm 1$ sublevels of the $5sep \ ^1P_1$ continuum. By adjusting the polarization vector of the ionizing laser perpendicular to that of

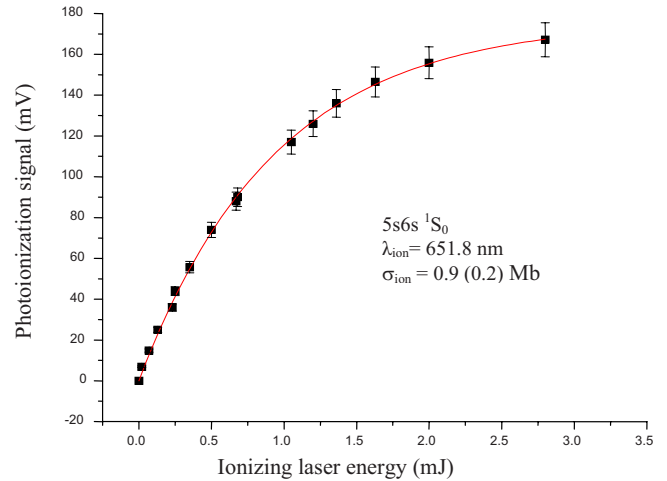


FIG. 2. (Color online) The photoionization signals from the $5s6s \ ^1S_0$ excited state plotted against the ionization laser energy. The solid line is the least-squares fit of Eq. (2) to the experimental data points, to extract the photoionization cross section σ (Mb) at the $5s$ ionization threshold.

the exciting laser, the accessible channels are $5sep \ J=1$ and $M_J = \pm 1$ and the corresponding cross section at the threshold has been determined as 1.1 (0.2) Mb. Subsequently, by adjusting the polarization vector of the ionizing laser at the magic angle 54.7° we have determined the photoionization cross section as 1.0 (0.2) Mb.

The cross section at the magic angle can be represented by the relation [29,36–39]

$$\sigma(\theta) = \cos^2 \theta \sigma^{\parallel} + \sin^2 \theta \sigma^{\perp}. \quad (3)$$

Here σ^{\parallel} and σ^{\perp} is the cross section as the polarization vectors of the ionizing dye laser is set parallel or perpendicular with respect to the excitation dye laser beam. Inserting the measured values of these cross sections (see above) this relation yields the cross-section value as 1.0 Mb, which is in excellent agreement with that of the experimentally determined value.

In a previous study [38] we presented two-photon excitation three photon ionization in strontium (one color two step) via the $5s6s \ ^1S_0$ intermediate state using a single dye laser at 653.77 nm which serves as an exciting as well as an ionizing laser. Since the third photon brings the atoms from the $5s6s \ ^1S_0$ state to nearly 45 cm^{-1} below the first ionization threshold therefore complete ionization of the excited atoms is not achieved which yields a lower value of the cross section (0.41 Mb). In the present work we have used two dye lasers, the first dye laser to saturate the intermediate state and the second laser at 651.87 nm to fully ionize the excited atoms. The photoionization cross section from the $5s6s \ ^1S_0$ state at the $5s$ threshold has been determined as 0.9 Mb. The overall uncertainty in the determination of photoionization cross section is about 20% which comes from fluctuations in the pump laser, energy measurement of the ionizing laser, transmission of the quartz windows, and in measuring the signal amplitude at the corresponding ionizing laser wavelength. The photoionization cross sections from the $5s^2 \ ^1S_0$ ground

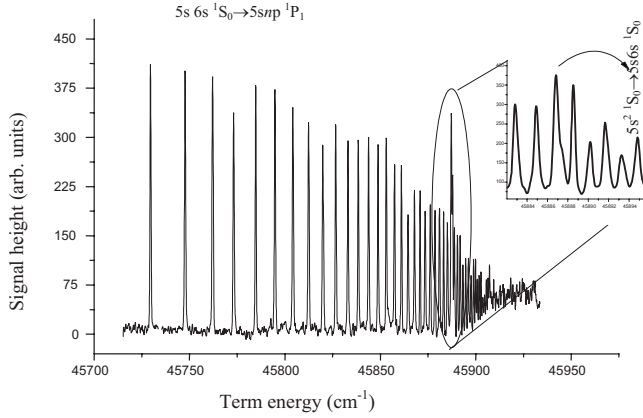


FIG. 3. A portion of the spectrum of strontium showing the $5snp\ ^1P_1$ ($26 \leq n \leq 73$) series excited from the $5s6s\ ^1S_0$ intermediate state. An inset in the figure shows the spectra recorded at a much lower laser intensity indicating significantly reduced intensity of the two-photon excitation three-photon ionization of the $5s^2\ ^1S_0 \rightarrow 5s6s\ ^1S_0$ transition.

state at the first ionization threshold range from 3 to 13 Mb (Yih *et al.* [40] and references therein) whereas a much lower value 0.9 (0.2) Mb has been obtained from the $5s6s\ ^1S_0$ state. The cross sections from the $5s5p\ ^1P_1$ and $5s5p\ ^3P_1$ excited states at threshold were reported as 11.4 (1.8) Mb and 10.7(1.7) Mb, respectively [19]. It will be interesting to measure the cross section from the $5s4d\ ^3D_2$ and $5s4d\ ^1D_2$ states to look for the ℓ dependence of the cross section in strontium.

In Fig. 3 we show the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ ($26 \leq n \leq 73$) Rydberg series recorded using two step excitation via $5s6s\ ^1S_0$ intermediate state. The pulse widths of both the dye lasers are 5 ns and the delay between the exciting and the ionizing laser is about 3 ns. The dye lasers are linearly polarized with parallel polarization vectors. The absolute term energies of the excited states are calculated by adding the excitation laser energy to the energy of the $5s6s\ ^1S_0$ ($30\,591.8\ \text{cm}^{-1}$) intermediate state [26,41]. The quantum defects for the observed series are calculated using the Rydberg relation

$$E_n = V_{\text{ion}} - \frac{R_y}{(n - \mu_\ell)^2}. \quad (4)$$

Here R_y is the mass corrected Rydberg constant of strontium $109\,736.6\ \text{cm}^{-1}$, E_n is the energy of the Rydberg level, V_{ion} is the ionization potential at $45\,932.09\ \text{cm}^{-1}$, and μ_ℓ is the quantum defect. A two-photon resonant three-photon ionization signal through the $5s6s\ ^1S_0$ intermediate state appears near the series limit at $45\,887.2\ \text{cm}^{-1}$ which causes irregularities in the line shapes. Since this line appears due to multiphoton excitation/ionization process, its signal height can be minimized by reducing the intensity of the dye laser. The corresponding spectrum recorded at a much lower intensity of the scanning dye laser is shown as an inset in Fig. 3.

The $5snp\ ^1P_1$ series in strontium has been extensively studied and the presently determined term energies are in excellent agreement with the earlier reported values

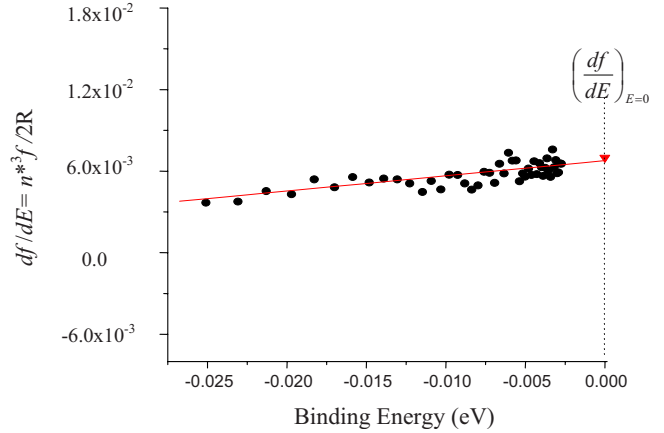


FIG. 4. (Color online) The experimentally measured oscillator strength distribution in the discrete region and at the $5s$ ionization threshold corresponding to the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ transitions.

[14,42–46]. In the present contribution we are concentrating only on the oscillator strengths of the transitions originating from the $5s6s\ ^1S_0$ excited state. The parameters required in Eq. (1) (λ_n , λ_{1+} , S_{1+} and S_n) to determine the oscillator strengths have been extracted from the recorded spectrum (see Fig. 3). The value of the photoionization cross section used in these calculations is determined under identical conditions, i.e., polarization vectors of both the dye lasers are kept parallel. In Table I we present the principal quantum number, excitation wavelengths, and extracted oscillator strengths of the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ Rydberg transitions. The experimentally determined oscillator strengths indicate a smooth decrease in f values as $n^{-2.75}$, ideally it should be n^{-3} for an unperturbed series. The deviation in the scaling law of the relative intensities may be attributed to the interaction of the $5snp\ ^1P_1$ series with the $4d5p\ ^1P_1$ level, which lies between the $5s7p$ and the $5s8p$ levels. This perturbation has been analyzed by Wynne and Armstrong [47]. The f values at $n=52$ and 53 are higher than that of the nearby members of the series (see Table I). This localized variation in intensities is attributed to the influence of the two-photon excitation three-photon ionization signal $5s^2\ ^1S_0 \rightarrow 5s6s\ ^1S_0$, which lies in the vicinity of these lines. Although its signal has been much reduced by decreasing the intensity of the scanning dye laser it still affects the line shapes and intensities of the nearby lines.

Parkinson *et al.* [10], Connerade *et al.* [14], and Mende and Kock [16] reported the f values for the $5s^2\ ^1S_0 \rightarrow 5snp\ ^1P_1$ Rydberg series. A comparison with the present data reveals that the f values for the $5snp\ ^1P_1$ series are higher if excited from the ground state $|\langle 5snp\ ^1P_1 | er | 5s^2\ ^1S_0 \rangle|^2$ than that of excited from the $5s6s\ ^1S_0$ level $|\langle 5snp\ ^1P_1 | er | 5s6s\ ^1S_0 \rangle|^2$. This is due to the fact that oscillator strength of a transition depends on the optical coupling between the states involved, i.e., $f_{ij} \propto |\langle \psi_j | er | \psi_i \rangle|^2$, therefore the f values for the $5s^2\ ^1S_0 \rightarrow 5snp\ ^1P_1$ and the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ transitions are anticipated to be different. We could not find any data in the literature to compare our experimental findings corresponding to the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ transitions.

To establish a connection between the oscillator strength distribution in the bound region and the differential oscillator

TABLE I. Optical oscillator strength of the $5s6s\ ^1S_0 \rightarrow 5snp\ ^1P_1$ transitions in strontium.

n	Wavelength (nm)	Oscillator strengths (units of 10^{-6}) ($\pm 20\%$)	n	Wavelength (nm)	Oscillator strengths (units of 10^{-6}) ($\pm 20\%$)
26	660.60	7.9	50	653.96	1.9
27	659.88	7.1	51	653.88	1.6
28	659.26	7.6	52	653.76	2.4
29	658.70	6.4	53	653.73	1.5
30	658.20	7.2	54	653.66	1.2
31	657.76	5.8	55	653.60	1.1
32	657.36	6.0	56	653.53	1.1
33	657.00	5.0	57	653.47	1.0
34	656.68	4.8	58	653.41	1.1
35	656.39	4.3	59	653.36	0.9
36	656.11	3.7	60	653.29	1.0
37	655.84	3.0	61	653.25	0.9
38	655.64	3.3	62	653.21	0.7
39	655.44	2.6	63	653.17	0.8
40	655.25	3.0	64	653.13	0.8
41	655.07	2.8	65	653.09	0.7
42	654.91	2.3	66	653.05	0.6
43	654.76	1.9	67	653.01	0.8
44	654.62	1.9	68	652.97	0.6
45	654.49	2.1	69	652.94	0.6
46	654.38	2.0	70	652.91	0.5
47	654.26	1.6	71	652.88	0.5
48	654.16	1.9	72	652.84	0.5
49	654.06	1.6	73	652.82	0.5

strength at threshold, we have used the following relation (Berkowitz [48]):

$$\frac{(n^*)^3 f_n}{2R} = \frac{df}{dE}. \quad (5)$$

Here, n^* is the effective quantum number, R is the Rydberg constant, and E is the photon energy. The spectral density of oscillator strengths is also related to the photoionization cross section as

$$\frac{df}{dE} = 9.11 \times 10^{15} \sigma(E) \text{ cm}^{-2}(\text{eV})^{-1}. \quad (6)$$

Using these relations and the measured value of the absolute photoionization cross section at threshold, we have determined the oscillator strength density in the bound region and the differential oscillator strength at threshold and their combined graph is shown in Fig. 4. The solid line is the linear fit of the oscillator strength distribution in the discrete region, indicating a smooth connection to the oscillator strength density across the ionization threshold; whereas the vertical dashed line represents the ionization threshold. The uncertainty in the determination of the oscillator strength is $\approx 20\%$, which is due to the measured photoionization cross

section at threshold ($\approx 18\%$) and the uncertainty in the measurement of the integrated signal intensity ($\approx 10\%$).

In conclusion, we have experimentally determined the oscillator strength distribution in the discrete region from the $5s6s\ ^1S_0$ excited state of strontium. The oscillator strengths of discrete states provide a sensitive test for atomic structure calculations and are useful for developing theoretical models involving electronic transitions induced by energetic radiation. There is room for further improvement in this method, particularly by the precise measurements of the photon flux and the energy of the ionizing laser, as there ought to be a linear relationship between the number of ions produced and the voltage drop across the thermionic diode. Further work in this direction is in progress in this laboratory.

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